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The potential of using biomass-based reducing agents in the blast furnace: A review of thermochemical conversion technologies and assessments related to sustainability

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ABSTRACT

Steel is one of the most important materials used in modern society. The majority of the steel produced today is based on the use of virgin raw materials such as iron ore and fossil-based energy sources such as coal and oil. Recently, emphasis has been laid on the possibility to replace part of the fossil-based energy sources in ironmaking with renewable, biomass-derived reducing agents (bio-reducers). Modern thermochemical conversion processes could be utilized to produce solid, liquid and gaseous bio-reducers from biomass feedstock. This paper provides an in-depth review, first of its kind, of the possibility to replace part of the fossil-based reducing agents with bio-reducers in the blast furnace ironmaking route. In the first part of the paper, blast furnace process and the role of the reducing agents are discussed. In the second part of the paper, thermochemical biomass conversion technologies and resulting product properties are reviewed. Chemical and physical properties of the bio-reducers are compared to fossil-based reducing agents. In the third part of the paper, the implications related to bio-reducer use are evaluated in separate system levels including unit process assessment (blast furnace), process integration opportunities, biomass availability, life cycle impact and economic evaluations. The fourth part of the paper is dedicated to the discussion of the future of bio-reducers in ironmaking applications. It is concluded that bio-reducers can contribute to more environmentally sound steelmaking. The most promising bio-reducer seems to be charcoal with its high coke replacement ratio in blast furnace and straightforward integration of slow pyrolysis by-products to heat and power applications.

Contents

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1. Introduction

Steel is one of the most important construction materials in modern society. Because of its versatile properties, steel is used in several applications. World crude steel production has increased significantly in the past decade. In 2012, the total production of crude steel was 1548 Mt [1]. The major part of crude steel, 64.4%, is produced via integrated blast furnace (BF)–basic oxygen furnace (BOF) route [2]. The production of steel via integrated BF–BOF-route is based on the use of virgin iron bearing raw material. The recycling rate of steel is high; nonetheless, scrap-based steel production through electric arc furnace (EAF) is restricted by the availability of scrap.

The demand for steel is increasing world-wide, making it inevitable to use virgin raw materials. The most important step in crude steel production is the blast furnace process, which is used to reduce the iron oxides and melt the iron into hot metal that is further refined into steel in oxygen-blown converters. The production of hot metal requires the use of carbon and hydrogen containing reducing agents that, usually, are fossil-based such as coke, coal, oil, natural gas and hot reducing gases.

Recently, renewable biomass-derived reducing agents have gained attention as possible substitutes for fossil-based reducing agents in the ironmaking blast furnace. Previous studies have concentrated on the assessment of blast furnace behavior with charcoal used as an auxiliary reducing agent, as well as the environmental burden and economics of charcoal use in ironmaking processes [3–6]. Although several forms of fossil-based reducing agents are used in the blast furnace process, of the biomass-derived reducing agent only charcoal has been extensively studied in the literature. However, current conversion technologies make it possible to produce solid, liquid and gaseous biomass-based reducing agents with physical and chemical properties similar to the fossil-based ones.

Several extensive reviews concerning thermochemical conversion routes have been conducted in recent years. Reviews of pyrolysis technologies to produce bio-oil and charcoal have been conducted e.g. by Bridgewater et al. [7], Bridgewater and Peacocke [8], Mohan et al. [9], Goyal et al. [10] and Vamvuka [11]. Reviews concerning the biomass gasification have been conducted e.g. by Zhang [12] and Kumar et al. [13]. The biomass conversion technology reviews have concentrated mainly on assessing the conversion of biomass into suitable forms for energy carriers such as transportation fuels or in chemical manufacturing.

In this paper, the suitability of thermochemical conversion technologies to produce biomass-based reducing agents (bio-reducers) is reviewed. Even though charcoal could replace small amounts of coal in metallurgical coke production [14], in this review only the option to replace tuyére injected reducing agents is considered. In the blast furnace, high hot strength of coke is required [15], which is difficult to maintain if charcoal is added to the coal blend. To understand the potential of biomass-based reducing agents in blast furnace ironmaking, an in-depth analysis of several factors is necessary. Currently, there is no reference available that thermochemical conversion technologies and resulting products would have been reviewed from the perspective of reducing agent use. This paper provides the first systematic insight into the use of renewable biomass in ironmaking applications by evaluating: (a) the current status of thermochemical conversion technologies, (b) properties of products from thermochemical conversion, (c) technological considerations at unit process level (blast furnace), (d) sustainability considerations at plant- and national level and (e) supply chain implications.

2. Fossil-based reducing agents in blast furnace ironmaking

2.1. Blast furnace technology

The blast furnace (BF) is the most used technology to produce hot metal for steelmaking purposes. The core of the process is to convert iron oxides into hot metal, which is done by utilizing carbon and hydrogen-based reducing agents. The BF is large counter-current metallurgical furnace where iron oxides and coke fed into the furnace from the top are moving downward, while reducing gases are moving upward. The reducing conditions in the furnace are created with the help of top-charged (coke) and tuyére injected (oil, pulverized coal, etc.) reducing agents. Coke and other carbon and hydrogen containing materials are combusted with the help of oxygen-enriched air that is blown from the tuyéres. The reducing gas flows upwards and reduces and melts the top-charged burden material. The total consumption of the reducing agents is below 500 kg/t hot metal in the most advanced blast furnaces, which is difficult to further decrease with current BF technologies [16].

Nomenclature

BAT	best available technology
BF	blast furnace
BFB	bubbling fluidized bed
BOF	basic oxygen furnace
CC	charcoal
CCS	carbon capture and storage
CFB	circulating fluidized bed
CV	calorific value
DRI	direct reduced iron
EAF	electric arc furnace
EBF	experimental blast furnace
EROI	energy return on investment
EU-ETS	European union emissions trading scheme
F-T	Fischer-Tropsch
GHG	greenhouse gas emissions
HHV	higher heating value
HM	hot metal
HRG	hot reducing gas
IT	innovative technology
LCA	life cycle assessment
LHV	lower heating value
NG	natural gas
NCV	net calorific value
PC	pulverized coal
PSA	pressure swing adsorption
SNG	synthetic natural gas
SA	sustainability assessment
TGR-BF	top-gas recycling blast furnace
ULCOS	ultra low CO_2 steelmaking

The use of coke is mandatory for the blast furnace process, as it acts as a support medium for the burden [17]. Several forms of fossil-based auxiliary fuels such as oil, coal and natural gas can be injected into the furnace with high velocity to provide heat and a reducing atmosphere. Use of these auxiliaries affects the behavior of the furnace and the consumption of coke.

2.2. Fossil-based reducing agents

Coke is the primary fuel and reducing agent in the blast furnace process. The amount of coke that is used in modern blast furnaces is around 350–400 kg/t hot metal, depending on the amount of auxiliary reducing agents used [18]. The metallurgical coke in the blast furnace performs three functions: (1) acts as a reducing agent, (2) provides energy to the process and (3) serves as a support medium for the burden material [19]. By injecting auxiliary reducing agents to the blast furnace, the amount of coke could be decreased to level of 200 kg/t hot metal [20,21]. However, some amount of coke is always needed in the blast furnace to act as a support medium.

Heavy oil (bottom oil) is used in the blast furnace as an auxiliary fuel. The best values in total reducing agent consumption (458.5 kg/t hot metal) in European blast furnaces with oil injection have been reached by the Finnish steel mill Ruukki, with 358.0 kg coke and 100.5 kg oil/t hot metal [18]. The injection of oil to the blast furnace became widely practiced method in the 1960s to enhance the productivity and control of the furnace. The oil crisis of the 1970s changed the paradigm towards coal injection [22]. The main advantage of oil injection is that it is an effective injectable reducing agent [23]. The heating value of the oil is 40.04 MJ/kg [24].

Pulverized coal is the most used auxiliary fuel in the iron blast furnace. High injection rates can be achieved with pulverized coal, which means that the coke amount can be decreased substantially. The chemical properties of the coal have a significant impact on the behavior of coal in the blast furnace. Good quality coal has high carbon content and a suitable volatile share. The ash amount decreases the heating value of the coal [17].

The amount of injected pulverized coal can reach over 200 kg/t hot metal. The highest reported amount is 235 kg/t hot metal while the amount of coke was 281 kg/t hot metal, total reducing agent consumption being 516 kg/t hot metal [18]. The typical composition of coke, pulverized coal and heavy oil is presented in Table 1.

Natural gas (NG) is also one of the reducing agents used in blast furnace ironmaking. The use of NG is common in countries where NG is inexpensive. The largest volume of injected natural gas has been reached in the USA, with 155 kg/t hot metal [25]. Higher injection rates of natural gas lead to local supercooling of the furnace hearth. The chemical properties of NG depend on the source and treatment of the gas. The volume fraction of methane can be in the range of 80–99%, 95% on average. Additionally, NG contains higher hydrocarbons, nitrogen, carbon dioxide and traces of other elements and compounds [26,27]. In blast furnace calculations, in many cases NG is assumed to be pure methane [28].

Radical changes can be achieved by introducing ***hot reducing gases*** (HRGs) into the blast furnace originating from different sources [29]. HRGs can be produced from coal gasification and introduced to the blast furnace [30]. Ziebik et al. [31] evaluated the utilization of COREX process off-gas, which has gone through Selexol separation process. Top-gas recycling blast furnace

Table 1
Composition of coke, pulverized coal and heavy oil [3,24].

Component (%)	Coke [3]	Pulverized coal [3]	Heavy oil [24]
C	88	80.6	87
H	0.35	4.35	10.45
O	0.5	5.35	ND
N	0.4	1.65	ND
S	0.6	0.45	2.01
Ash	9.63	10.89	0.02
CaO in ash	2.37	1.49	ND
SiO_2 in ash	56.25	57.15	ND

Table 2
The composition and heating values of gaseous fossil-based reducing agents [20,25,30,33].

	Natural gas	HRG 1 [30]	HRG 2 [30]	HRG 3 [20]	HRG 4 [33]	HRG 5 [25]	HRG 6 [25]
CO (%)	–	51.5	72.0	71.7	62.2	22.6	60.0
H_2 (%)	–	44.0	25.0	14.9	26.7	71.0	33.0
CH_4 (%)	100	–	–	–	1.5	3.0	3.0
CO_2 (%)	–	3.0	1.5	3.2	5.2	3.0	4.0
N_2 (%)	–	0.0	0.0	10.3	3.0	1.0	–
H_2O (%)	–	1.0	1.5	–	1.5	–	–
LHV (MJ/Nm^3)	35.8	11.2	11.8	10.7	11.3	11.5	12.2

Here the chemical composition and calculated lower heating values of gaseous reducing agents are presented. It can be derived from the data that wide range of hot reducing gases can be injected into blast furnace.
HRG = hot reducing gas.
LHV = lower heating value (MJ/Nm^3).

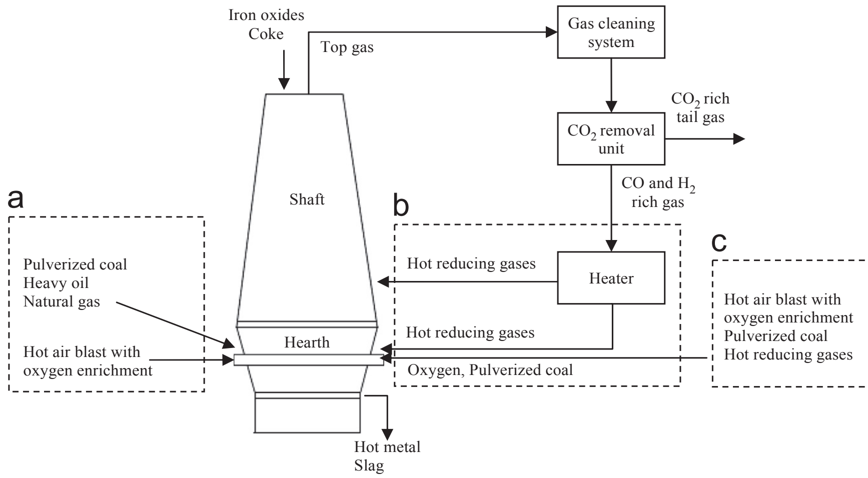


Fig. 1. Different blast furnace configurations with fossil-based reducing agents. BF is flexible technology concerning the fuel use. Liquid, solid and gaseous reducing agents can be used.

(TGR-BF) is an emerging concept that has not yet been commercialized but is under intense development stage e.g. in the scope of ULCOS program [32]. In the TGR-BF process, the top gas of the blast furnace is re-circulated back to the furnace after stripping CO_2 from the gas [5]. The top gas recycling concept necessitates the use of oxygen instead of air, to burn carbon in the lower part of the furnace and to produce reducing gases and heat. Usually, the TGR-BF concept is accompanied with coke consumption of around 190–200 kg/t hot metal, pulverized coal rate of 170–180 kg/t hot metal and hot reducing gases of 600 m^3/t hot metal. The fossil carbon consumption in TGR-BF is around 300 kg/t hot metal [20].

The composition of hot reducing gases differs in relation to the share of H_2 and CO in the gas mix. However, the total share of (H_2+CO) should be in the range of 90% of the gas mix volume. High hydrogen content of hot reducing gases would be beneficial for the process [25]. In Table 2, the composition and heating values of HRGs, evaluated as possible reducing agents in the blast furnace, are presented. The H_2/CO volume ratio of hot reducing gases that can be injected into the blast furnace ranges from 0.2 to 3.1. The calculated lower heating values (LHV) of HRGs range from 10.7 to 11.8 MJ/m^3, while the LHV of NG is 35.8 MJ/Nm^3.

Fig. 1 depicts different configurations with alternative reducing agents in the blast furnace. Box (a) illustrates a conventional case, where fossil-based reducing agents; coal, oil or NG are used as tuyére injectants and coke is charged from the top of the furnace. Box (b) illustrates a case where top gas is conditioned with CO_2 removal and recycled back to the furnace as hot reducing gas. Box (c) illustrates a scenario where HRGs are produced e.g. from coal gasification and injected solely or with co-injection with pulverized coal [25,30]. HRGs can be injected into the blast furnace at the bottom of the furnace (hearth zone) or at the lower part of the shaft.

Table 3 summarizes the reducing agent use with different operating options. Coke is always needed in the blast furnace, but the injectants can be varied. Auxiliary reducing agents have varying potential to decrease the coke consumption. The numbers presented are based on the actual performance of operating blast furnaces and on mathematical modeling and simulation studies [18,25]. The consumption figures are indicative as the reducing agent need is influenced by several factors.

The lowest total reducing agent use has been achieved with oil injection; however, the amount of coke has been rather large. The amount of HRG with composition of HRG 6 in Table 2 seems large. However, HRG replaces major part of the hot air that is needed to burn carbon from coke and from auxiliary fuels.

Table 3
Total reducing agent consumption in the blast furnace [18,25].

		Main auxiliary fuel			
		Oil [18]	**PC** [18]	**NG** [25]	**HRG** [25]
Coke	kg/t HM	358.0	280.9	310	365
Injectants					
Heavy oil	kg/t HM	100.5	0.9		
Pulverized coal	kg/t HM		235.1		
Natural gas	kg/t HM			155	
Hot reducing gases	kg/t HM				529.6
Total reductants	kg/t HM	**458.5**	**516.9**	**465**	**894.6**

It summarizes the total reducing agent consumption with different auxiliary fuels injected from the lower part of the blast furnace.
HRG = hot reducing gas.
NG = natural gas.
PC = pulverized coal.

3. Production of reducing agents from biomass by thermochemical conversion technologies

3.1. Biomass properties

Several biomass feedstocks have been found suitable for producing solid reducing agents for iron and steelmaking purposes. The studied biomass feedstock include straw, saw mill waste, planted wood-species and forest residues such as thinnings, stumps and roots [3,4,34].

Biomass comprise mainly of carbon (C), hydrogen (H), oxygen (O), nitrogen (N) and sulfur (S). The share of carbon in the wood is around 50 wt% (dry matter, dm), depending on the wood species and the part of the wood (bark and stem). The carbon content is low compared to fossil fuels such as coal, coke or oil that are used in iron and steelmaking. The share of oxygen in the wood is around 40 wt% (dm). The existence of oxygen in the biomass decreases its energy content. The fixed carbon content in the biomass is low, around 10–16 wt% (dm), while volatile matter is 84–88 wt% (dm) and ash (A) content 0.4–0.6 wt% (dm). The sulfur content in the wood biomass is low around 0.01–0.1 wt% (dm) [35]. The low sulfur content is advantageous for blast furnace ironmaking [36]. A thorough analysis of principal components (C, H, O, N, S and A) in different biomass species and assortments has been conducted by Tao et al. [37]. According to their statistical

analysis on extensive amount of species (144), it was concluded that CHO contents in different biomass species and assortments are less variable than other variables (NSA contents). Woody materials were more homogenous in composition than herbaceous biomass such as straw and fruit residues and had lower ash content. Wood was considered as a high quality, but straw a low quality fuel [37].

The ash content of woody biomass is usually low, between 0.5% and 2% in most wood species and varies in different parts of the wood [35,38]. Whole tree chips have lower ash content compared to bark [35]. The ash content of herbaceous biomass and agricultural residues is typically higher than in woody biomass, upto almost 40% [38,39]. Woody biomass ash is generally rich in calcium (Ca) and potassium (K) and herbaceous biomass ash is rich in silicon (Si), K and chlorine (Cl) content [38]. The alkali metal content of the biomass is important for blast furnace behavior. Alkalis increase coke consumption and the possibility of scab formation that disturbs the process [40].

The heating value of biomass is low compared to fossil fuels. The net calorific value (NCV), or lower heating value (LHV) of wood biomass dry matter is between 18.3 and 20 MJ/kg [35]. This assumes zero moisture content. Higher heating value (HHV) of biomass fuels is generally between 14 and 23 MJ/kg on dry basis [41]. Herbaceous materials have typically lower HV than woody materials, mainly because of higher ash content and lower carbon content [38,41]. The moisture of the biomass is a critical factor for its use. The moisture of green wood can be as high as 60% [35], whereas the moisture content in agricultural residues is generally much lower [42]. The thermochemical conversion technologies require low moisture content feedstock [39]. The moisture affects the energy need of the conversion process, but also has an influence on the product quality.

The bulk density of biomass affects the economics of the use, due to transportation and storage costs. Bulk density influences also the conversion process behavior. The dry and ash free bulk volume of wood chips is 4.4–5.6 m^3/t [39]. Pre-treatment such as pelletizing results in more efficient bulk volumes (1.6–1.8 m^3/t, dry and ash free, daf).

3.2. Pathways to produce reducing agents

The production of reducing agents from biomass feedstock has been acknowledged by several authors [19,43,44]. In the literature, the most emphasis is placed on the assessment of solid bioreducer, i.e. charcoal. The most studied biomass feedstock related to ironmaking purposes is wood. In Brazil, for example the use of charcoal derived from wood-based feedstock is common practice in ironmaking [45,46].

There are several thermochemical biomass conversion processes where the properties of biomass are enhanced for use. The main thermochemical conversion routes for biomass are presented in Fig. 2. Apart from the solid reducing agents, also liquid and gaseous reducing agents can be produced from biomass feedstock. The conversion technologies produce valuable byproducts that can be utilized in several applications such as chemical industry and electricity production [4].

Firstly, the pre-treatment processes are required to set acceptable particle size and moisture of the feedstock. The main primary conversion technologies evaluated in this paper are slow pyrolysis, fast pyrolysis and gasification. The primary products are charcoal, bio-oil, syngas and synthetic natural gas through methanation. In addition, Mathieson et al. [47] have evaluated the performance of torrefied softwood as blast furnace injectant, however, found it unsuitable.

For optimal blast furnace performance, the key factors for the reducing agents are sufficient energy content and the ability to form reductive atmosphere in the furnace conditions. The typical product weight yields for pyrolysis of wood are presented in Table 4 [48].

3.3. Charcoal from slow pyrolysis

Pyrolysis can be described as thermal decomposition of material in the absence of oxygen. Slow pyrolysis is a process where modest heating rates are used. Lower processing temperature and longer residence times favor the production of charcoal. The temperature employed in the production of charcoal is around

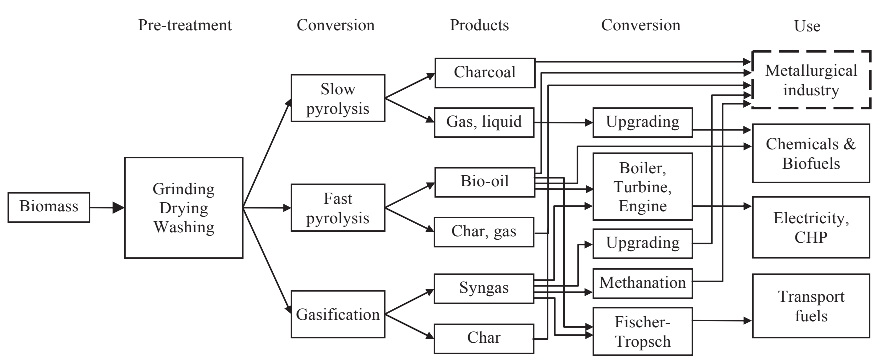


Fig. 2. Pathways to produce reducing agents for metallurgical industry from biomass by thermochemical conversion.

Table 4
Typical product weight yields (db) of pyrolysis of wood [48].

Mode	Conditions	Liquid	Solid	Gas (%)
Fast	~500 °C, short hot vapor residence time ~1 s	75	12 char	13
Intermediate	~500 °C, hot vapor residence time ~10–30 s	50 in 2 phases	25 char	25
Carbonisation (slow)	~400 °C, long vapor residence hours to days	30	35 char	35
Gasification	~750–900 °C	5	10 char	85
Torrefaction (slow)	~290 °C, solids residence time ~10–60 min	0 Unless condensed, then upto 5	80 solid	20

500 °C and the residence time can be several hours [49,50]. Charcoal could replace pulverized coal injection in the blast furnace, but there are some properties that have to be monitored [3,4,47,51].

3.3.1. *Properties of charcoal compared to pulverized coal*

The properties of charcoal and resulting side-products are highly dependent on processing conditions. In Fig. 3., a yield of char, gas, condensed liquids and tar from slow pyrolysis of wood are presented accompanied with charcoal carbon content [52]. With increasing temperature, the yield of charcoal decreases and carbon content increases. High temperatures favor the formation of gases.

The charcoal yield of 30–35% can be achieved with final temperatures between 450 and 550 °C, with carbon content ranging from over 80% to almost 90% [53]. The increase in pressure from 0.1 MPa to 1.0 MPa increases the yield of charcoal from hard and soft wood on average from 30% to 35% [54]. Other factors that affect the properties of the final charcoal are type, nature and composition of feedstock [52]. Especially important from the metallurgical use point of view are the ash content and the composition of ash. Vapor residence time, heating rate and particle size have also an effect on the formation of solid product [49].

The chemical properties of the charcoal are important for the blast furnace process behavior. In order to generate suitable gas flow patterns to the blast furnace, the amount of volatiles in the injected char should be optimized [17]. This is relatively straightforward to achieve with charcoal, as the carbonization conditions can be controlled to required carbonization degree or fixed-C content. In Table 5, three fossil-based pulverized coal samples and four types of charcoals are compared. The composition of pulverized coals 1–2 (PC 1, PC 2) are taken from Ref. [3] and the composition of pulverized coals 3–4 (PC 3, PC 4) are taken from Ref. [55]. The composition of charcoal 1 (CC 1) is taken from Ref. [3], charcoal 2 (CC 2) from Ref. [55] and charcoal 3–4 (CC 3 and CC 4) from Ref. [56]. CC 1 is produced from eucalyptus in laboratory oven, CC 2 is from industrial producer in Brazil, and CC 3 and CC 4 are hardwood charcoals from the same industrial producer. The carbon content of the pulverized coals and charcoals are similar. The amount of volatiles depends on the carbonization degree.

The chemical properties of the charcoal are well comparable to pulverized coal. The carbon content of the charcoal can be even higher than with pulverized coals. The sulfur content of the charcoal is low compared to fossil-based reducing agents (PC∼0.5, heavy oil ∼2.0). Sulfur is a harmful element in hot metal and requires intensified procedures in subsequent processes.

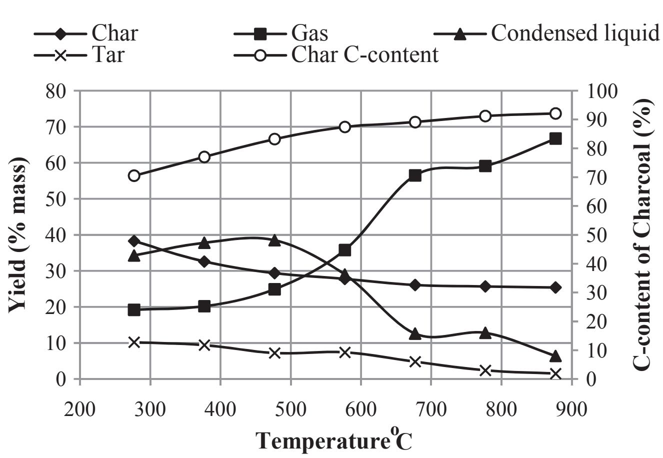


Fig. 3. Yields of slow pyrolysis products and carbon content of charcoal. It shows the relationship between pyrolysis temperature and yield of char, gas, condensable gases and tar. C-content is the total carbon content of the char, which increases when temperature increases.
(data from [52])

Table 5
Properties of pulverized coal and charcoal [3,55,56].

Coal	Proximate analysis (wt%)			Ultimate analysis, daf (wt%)				
	Volatile matter daf	Fixed C daf	Ash db	C	H	O	N	S
PC 1 [3]	30.40	ND	7.49	80.0	4.5	6.49	1.2	0.32
PC 2 [3]	8.60	ND	10.27	82.80	3.3	2.31	0.9	0.42
PC 3 [55]	27.7	72.3	9.5	86.4	4.9	6.0	2.1	0.6
PC 4 [55]	18.5	81.5	10.3	89.3	4.3	3.5	2.0	0.5
CC 1 [3]	18.82	80.69	0.6	88.26	2.71	8.42	0.21	0.03
CC 2 [55]	21.8	78.2	4.6	83.7	3.1	12.0	1.1	0.1
CC 3 [56]	9.46	88.63	1.91	91.65	2.42	3.64	0.33	<0.05
CC 4 [56]	13.31	82.39	4.30	86.38	2.30	6.47	0.50	<0.05

Proximate and ultimate analyses for four pulverized coals and four charcoals are presented. It can be seen that pulverized coal (PC) and charcoal (CC) have quite similar properties. The properties of the charcoals can be influenced by the selection of carbonization temperature. The ash content of PC is higher than that of CC.
CC=charcoal.
db=dry basis.
daf=dry and ash free.
PC=pulverized coal.

Table 6
Chemical analyses of the ashes of pulverized coals and charcoals [55,56].

Ash analysis (db)	PC 3 [55]	PC 4 [55]	CC 2 [55]	CC 3 [56]	CC 4 [56]
SiO_2	46.00	50.14	28.46	3.82	21.47
Al_2O_3	25.19	26.73	3.96	1.17	4.84
Fe_2O_3	14.61	9.03	1.95	1.14	4.40
TiO_2	1.24	1.33	0.23	0.15	0.28
P_2O_5	1.64	1.55	1.29	4.04	2.58
CaO	4.12	3.91	39.46	53.04	38.45
MnO	0.14	0.09	0.62	ND	ND
MgO	1.69	1.53	4.32	11.45	4.05
SO_x	1.20	0.78	1.91	4.67	2.62
Na_2O	0.21	0.41	0.12	0.47	1.62
K_2O	1.22	1.04	2.40	4.32	9.28
BaO	ND	ND	ND	1.03	0.42
SrO	ND	ND	ND	0.28	0.18
NiO	ND	ND	ND	0.05	<0.007
V_2O_5	ND	ND	ND	0.20	<0.020

Here the chemical analyses of two pulverized coals and three charcoals are presented. PC ash is acidic in nature, whereas CC ash is basic.
CC=charcoal.
db=dry basis.
PC=pulverized coal.

The ash amount and chemical analyses of injected chars influence the blast furnace process. The ash amount of pulverized coals is in the range of 10%, whereas the ash amount in the charcoals is a couple of percentages at maximum. The ash chemistry of coals differs considerably between fossil-based coals and wood-based charcoals. In Table 6, the chemical analyses of the ashes of two pulverized coals and three charcoals are presented [55,56]. The acronyms correspond to the ones used in Table 5, to relate to the same coal sample. The pulverized coals have a high content of SiO_2 and Al_2O_3, whereas the charcoals contain elements that increase basicity (e.g. Ca and Mg).

The basicity of the ash has advantageous effects to the formation of slag, which is used in the blast furnace to collect unwanted elements from the hot metal. The acidic nature of the charcoal

reduces the need of lime or limestone addition, thus decreasing the amount of slag. The reduced amount of slag in the blast furnace process increases productivity [3,36].

3.3.2. Slow pyrolysis technologies

There are several technologies to produce charcoal ranging from traditional clay charcoal pits to continuous processes that can be efficiently controlled. The charcoal yield with traditional earth pits and mounds is low (> 10%). Higher yields can be achieved with brick, concrete and metal kilns (20–25%). Batch process retorts can achieve yields of 30% and continuous processes such as Lambiotte retort can reach 30–35% yields [57]. The heat required by the slow pyrolysis process is usually supplied by one of the three methods: (1) directly, by burning part of the raw material under controlled air flow, (2) directly, by directing part of the combustion gases from pyroligneous vapor burning into the reactor, and (3) indirectly, by heating reactor outside with flue gases [58]. Heat carrier can also be other than flue gases, e.g. sand [57].

The development of solid product processes i.e. to produce char from biomass has accelerated in recent years due to application of biochar for soil remediation [59] and torrefied biomass for energy production [60,61]. Pyrolysis technologies have also been used in waste treatment applications [62]. Microwave-assisted pyrolysis technology has also been proposed for the production of solid chars [63,64]. The same type of technologies could be used for producing charcoal for reducing purposes. Charcoal production for ironmaking has achieved a large scale e.g. in Brazil. In 2010, hot metal production in mini blast furnaces based on charcoal was 5.8 Mt, whereas prior to the economic crises the level was 9.4 Mt in 2007 [45]. The biggest production units in the world for charcoal production are around 25,000 t charcoal per year. These kinds of capacities can offer e.g. Lurgi continuous retort process and Degussa batch process [1,58]. Commercial slow pyrolysis plant (MTK) exists in Japan that processes 100 t biomass per day in a rotary kiln [65]. The majority of the slow pyrolysis technologies are small-scale and applicable to produce biochar and energy from agricultural waste and forest residues (e.g. Pro-Natura and Pyreg, [66,67]). Usually integrated steel plants are large and iron blast furnaces use significant amounts of reducing agents. Therefore, the production of charcoal should be in an industrial scale to make it practical. The production of charcoal could be centralized or decentralized, but the utilization of slow pyrolysis by-products should be taken into account [44].

There are four major steps in continuous slow pyrolysis process: pre-treatment, pyrolysis, solids removal and heat generation [68]. A simplified process flow chart for charcoal production by slow pyrolysis is presented in Fig. 4. In a study by Brown et al. [68], pyrolysis is conducted in a fluidized bed reactor; however, other process technologies are more of a common.

Roberts et al. [69] state that 50% of the feedstock energy coming into the slow pyrolysis process via feedstock goes to the solid product, i.e. charcoal. The overall efficiency from feedstock to available heat is reported to be 37%. According to Brown et al. [68], the mass flow rate on dry basis is distributed as following: 36% biochar and 64% pyrolysis gas. With the assumption that the feedstock heating value (db) is 19 MJ/kg and the biochar heating value (db) is 30 MJ/kg, the energy fixed to charcoal constitutes to 57%, while the pyrolysis gases constitute to 43%. The energy value of pyrolysis gas calculated from the balance is 12.8 MJ/kg on average, considering no losses. The distribution of the energy depends on the carbonization degree, yield and technology applied.

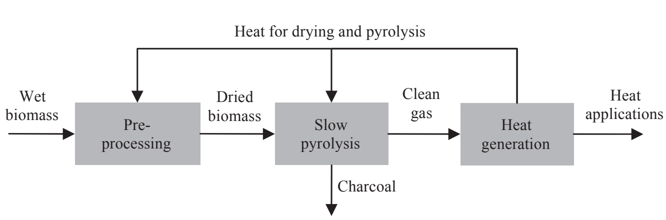


Fig. 4. Simplified process flow chart of charcoal production with heat and power application . It illustrates in simplified manner how slow pyrolysis can be applied to produce charcoal and heat and power. Slow pyrolysis is self-sustaining and only about 50% of the incoming feedstock energy is in charcoal. Rest of the energy goes to liquids, gases and losses.
(modified from [68])

3.4. Bio-oil from fast pyrolysis

Fast pyrolysis is a thermochemical biomass conversion process aiming at maximum yield of a liquid product in the absence of oxygen. In the fast pyrolysis process, higher heating rates are used and quick cooling of volatile products is employed. The temperature in fast pyrolysis is modest (around 500 °C), but the vapor residence time is short. The main benefits of producing liquids from biomass are increasing energy intensity, storability and transportability of the product. The fast pyrolysis process requires good temperature control and small particle size [49,70]

The yield of the bio-oil can be as high as 70–80 wt% on dry feed basis. This includes also the water content, meaning that the yield of organics is around 60 wt% on dry feed basis. Usually, the by-products from the fast pyrolysis (solid char 15–25 wt% and non-condensable gases 10–20 wt%) are used within the system. The fast pyrolysis process requires that the biomass feedstock is dried below 10% moisture. This procedure minimizes the share of water in the primary product, which is oil. The feed should also be grinded to small particles (around 2 mm for fluid bed reactors) to enable rapid pyrolysis reaction, separation of solids, and collection of the liquid product [7,9,48,49]. Bio-oil is not a product of thermodynamic equilibrium, but short reactor times and quick cooling are used in the production [71].

3.4.1. Properties of bio-oil compared to heavy oil

Bio-oils are free-flowing organic liquids, which comprise of different sized oxygenated compounds. The properties of the bio-oil resemble more the properties of biomass than that of petroleum oils. The oxygenated compounds found in bio-oil are a result of depolymerization and fragmentation reactions of cellulose, hemicellulose and lignin [71,72]. The maximum yield of the bio-oil is achieved in the temperature range of 450–550 °C almost for any type of biomass [7,48]. As in the case of slow pyrolysis, also fast pyrolysis conditions influence the elemental and chemical composition of the product.

The water content in the bio-oil is high. According to the review of Butler et al. [73], the water content in the final bio-oil product produced with different technologies and from different feedstocks can be as high as 51%. The bio-oil water content is affected by the original moisture of the feedstock and the dehydration reactions taking place during pyrolysis, which makes the drying of the feedstock an important factor [72]. The distillability of bio-oil is poor, which makes the removal of water from the product almost unattainable [48]. The oxygen content of bio-oil is usually in the range of 35–40 wt%, and can be found in most of the over 300 compounds identified in bio-oils [72]. The properties of bio-oil from wood and microalgae feedstock compared against heavy fuel oil are summarized in Table 7 [72,74].

For use in a blast furnace, the high water and oxygen content of the bio-oil make it less suitable as a reducing agent. The heating value of bio-oil produced from wood-based feedstock is less than half of the heating value of heavy oil used in blast furnaces as an energy source and reducing agent. Compared with bio-oil from

Table 7
Comparison of the properties of heavy fuel oil and bio-oils [72,74].

Properties	Heavy fuel oil	Bio-oil	Autotrophic microalgae	Heterotrophic microalgae
Carbon content (%)	85	54–58	62.07	76.22
Hydrogen content (%)	11	5.5–7.0	8.76	11.61
Oxygen content (%)	1.0	35–40	19.43	11.24
Nitrogen content (%)	0.3	0–0.2	9.74	0.93
Ash content (%)	0.1	0–0.2	ND	ND
Moisture content (%)	0.1	15–30	–	–
pH	–	2.5	ND	ND
Specific gravity	0.94	1.2	1.06	0.92
HHV (MJ/kg)	40	16–19 as produced	30 (moisture free)	41 (moisture free)
Viscosity (cP)	180 (at 50 °C)	40–100 (at 50 °C)	100 (at 40 °C)	20 (at 40 °C)
Solids (wt%)	1	0.2–1	ND	ND

Here the properties of bio-oils and heavy fuel oil are compared. It can be seen that the bio-oil derived from the wood feedstock has lower quality for use in blast furnace than bio-oil derived from microalgae, which has nearly comparable properties with heavy fuel oil.
HHV = higher heating value (MJ/kg).

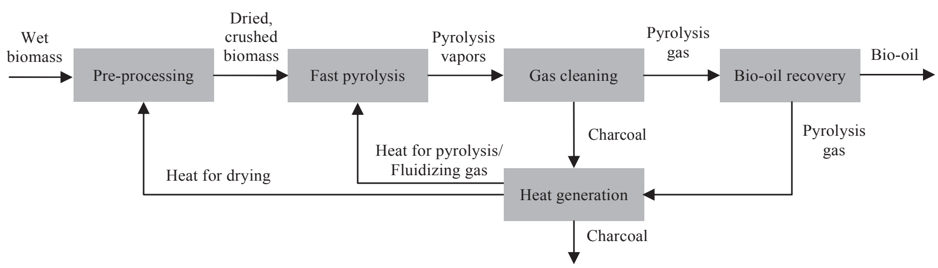


Fig. 5. Simplified process flow chart of fast pyrolysis process. Fast pyrolysis process is more complicated than slow pyrolysis, with gas cleaning and bio-oil recovery steps. (modified from [68])

wood, bio-oil from heterotrophic microalgae has much higher heating value, nearly comparable to fossil-based heavy oil. It seems that bio-oils from microalgae have higher quality for use in metallurgical purposes than those from lignocellulosic materials [74]. The main advantage of the microalgae compared to many other biomass feedstocks is the high oil productivity, which exceeds the oil productivity of oil crops. Additionally, microalgae grow extremely rapidly, which is why it is estimated to be the number one source of biomass to replace fossil-based transportation fuels [75].

The viscosity of bio-oil ranges from 35 to 1000 cP at 40 °C, and depends greatly on feedstock and process conditions. However, the viscosity of bio-oil decreases rapidly when the temperature is increased, more rapidly than with petroleum oils [72]. The viscosity of forest residue derived bio-oils produced in the temperature range of 480–520 °C with transport bed reactor was 20 cP for stored (brown) residues and 29 cP for green residues (at 40 °C) [76], which are rather low. The high viscosity reported by several authors for bio-oils derived from biomass would not be a severe problem to blast furnace process. A modest increase in temperature decreases the bio-oil viscosity to a suitable range. The heavy oil viscosity at injection temperature (around 200 °C) is usually between 20 and 30 cP, which can be achieved with bio-oils.

3.4.2. Fast pyrolysis technologies

Fast pyrolysis technologies that can be used for producing the bio-oil have been reviewed in the work of Bridgewater and Peacocke [8] and more recently by Bridgewater [48], Butler et al. [73] and Meier et al. [64]. There are numerous technologies available, the most typical being bubbling fluid bed, circulating fluid bed, transported bed, rotating cone, ablative pyrolysis and vacuum pyrolysis technologies [48,73,77]. In the fast pyrolysis process, there are basically five major processing steps to convert biomass into liquid bio-oil; pretreatment, pyrolysis, solids removal, oil recovery and heat generation [48,68].

Pre-processing of the biomass include the drying of the biomass to a moisture content of below 10% and grinding the biomass to particle size of 2–4 mm. In the pyrolysis reactor, the biomass is subjected to heat that is supplied in one of the two main ways. The heating of biomass can be based on gas-solid or solid-solid heat transfer. Bubbling fluid bed, circulating fluid bed, transported bed and rotating cone technologies utilize mainly solid-solid heat transfer from fluidizing solid. Entrained flow reactor utilizes gas-solid heat transfer from the hot gas. In the ablative reactor type, heat is led to the biomass through the reactor wall [7,48,77]. Solids removal means the separation of char and fluidizing solid that go through the pyrolysis reactor with cyclones [70]. Bio-oil recovery can be done with indirect heat exchangers and electrostatic precipitators [68]. The recovery of bio-oil should be done immediately after the vapors exit the pyrolysis reactor. The high temperature vapor continues the cracking reactions, which reduce the yield of organics in the bio-oil [8,78]. The process flow sheet of fast pyrolysis for bio-oil production is presented in Fig. 5. The utilization of by-products (char and non-condensable gases) depend on the technology but, basically, they are used as fuel inside the system. However, if alternative fuel is available, the charcoal could be exported from the system [48].

Several fast pyrolysis technologies are in the process of commercialization, some already in operation [64,73,79]. The largest bio-oil production units are around 200 t/d dried biomass. With the 70 wt% bio-oil yield, the yearly production would be 46 200 t bio-oil (330 operating days/year assumed). The process is based on a bubbling fluidized bed provided by Canadian Dynamotive [48,73]. Bio-oil production can also be seen as a method for densification of biomass to be further refined to transportation fuels in centralized manner [80]. Production units with biomass input of 1.08 Mt are seen optimal for bio-oil production [81].

One interesting option could be the utilization of charcoal by-product of fast pyrolysis in blast furnace as a reducing agent. The yield of charcoal in fast pyrolysis is around 15–25 wt%, depending on the feedstock [9]. Mullen et al. [82], have conducted experimental fast pyrolysis on a laboratory scale and analyzed different pyrolysis products. They used corn cob feedstock in their pyrolysis experiment with 500 °C temperature and bubbling fluidized bed reactor. The properties of the charcoal by-product were as follows: carbon 77.6%, hydrogen 3.05%, nitrogen 0.85%, sulfur 0.02% and oxygen 5.11%. The ash content was 13.4% and corn cob moisture content 6.8%. The carbon content and the higher heating value (30.0 MJ/kg) are in the range of suitable blast furnace reducing agent feed. The ash content of the by-product charcoal is high, but the ash content of pulverized coal can also be well above 10% [3].

3.5. Syngas from the gasification of biomass

Gasification as a term is usually referred to thermochemical conversion of biomass to gaseous form by using a gasification medium air, oxygen, steam, nitrogen, carbon dioxide or a combination of these [13]. Essentially, gasification is partial oxidation of the biomass [83]. Depending on the gasification agent, the method of operation and the process conditions, three product gas qualities can be produced. The calorific values (CV) of the three product gas from biomass gasification are [83]:

Low CV 4–6 MJ/Nm^3 (air and steam/air)
Medium CV 12–18 MJ/Nm^3 (oxygen and steam)
High CV 40 MJ/Nm^3 (hydrogen and hydrogenation)

Gasification products are used in several applications. The low calorific value gas can be used directly as a fuel gas in turbines and gas engines [84]. Syngas from oxygen or steam gasification can be used to produce e.g. methanol, hydrogen and Fischer–Tropsch liquids [85–87]. Gasification temperature is usually in the range of 600–1000 °C, but higher temperatures are used with certain reactor types. Heat required to raise the temperature can be supplied either directly or indirectly [13].

3.5.1. Properties of syngas compared to hot reducing gases

According to the literature, the main requirement of hot reducing gas utilization in ironmaking applications is sufficient reducibility. This means that the share of $CO+H_2$ in the gas should be as high as possible, in the range of 90% [25]. This requires the use of oxygen as gasification agent or the use of allothermal technology. Several variables influence the product gas properties. The moisture of the feedstock should be lower than 30% to ease ignition. The moisture also lowers the quality of the product gas by lowering its calorific value. The ash content of the biomass should be low as well. In many cases, the temperatures in the reactor exceed the melting point of the ash leading to slagging problems [83].

3.5.2. Gasification technologies

There are several technologies to produce syngas from biomass. Based on the method of heat supply, they can be divided into allothermal (indirect) and autothermal (direct) technologies [12]. For the purpose of use as a reducing agent, the syngas should be nitrogen free. This has been addressed with direct reduced iron (DRI) production [43]. Nitrogen dilutes the gas and, when used as a reducing agent, gas volumes would increase significantly. The presence of nitrogen would also increase the size of the gasifier and other equipment [84].

Circulating fluidized bed (CFB) and bubbling fluidized bed (BFB) reactors are typically used for autothermal gasification [12]. Fluidized bed gasifiers have been used for coal gasification for years. The use of CFB in paper industry for gasification of forest residues is of a common. Gasifiers can be operated at elevated pressures [83]. Autothermal gasifiers need oxygen, which raises investment costs [12].

Allothermal gasification is usually conducted in a dual bed gasifier. Göransson et al. [88] have conducted an extensive review on the recent developments in indirect gasification technologies with dual fluidized-bed gasifiers. The basic operating principle of allothermal dual-bed gasifier is that one bed is operated with steam to gasify the biomass into syngas at temperatures of 700–900 °C. The other bed is operated with air to burn the char to produce heat for the gasification. The heat is transported from combustion bed to gasification bed with solid heat carrier. The bed material and leftover char are transported back to the combustion bed. The investment costs are modest compared to autothermal designs, and the technology is scaleable upto 200 MW [12].

Entrained flow gasification technologies are used widely in coal gasification. The capacities of entrained flow gasifiers in coal gasification can be several hundred MW_{th}. Entrained flow gasification requires small particle size ($<$ 0.1–0.4 mm), which requires extensive milling, thus resulting in high electricity consumption [83,89]. Entrained flow gasifiers operate at high temperatures and carbon conversion rate is high with a low residence time, enabling high capacities [90]. In Table 8, indicative syngas compositions from different gasification reactor systems are presented [12].

Syngas produced with gasification needs to be cleaned and CO_2 content decreased before it can be utilized as a reducing agent. The cleaning of synthesis gas does not require as intensive purification as in the case of using it e.g. for Fischer–Tropsch synthesis. The emphasis is in the removal of solids that might disturb the injection of reducing gas to blast furnace. The synthesis gas contains considerable amounts of hydrocarbons that have to be converted into CO and H_2 when producing Fischer–Tropsch liquids [84]. In the case of blast furnace injection with hot reducing gases derived from coal gasification, a 3% share of CH_4 has been found to be acceptable [25]. The amount of hydrocarbons could be even higher; however, it would further require heat for the splitting and combustion of the hydrocarbons. High content of H_2 and CH_4 in the hot reducing gas will lead to high energy losses; however, proper control of the process makes it possible to utilize these gases [91]. Reforming of the synthesis gas can be done by several means. For example steam reforming with nickel catalyst and autothermal reforming can be used to convert hydrocarbons into CO and H_2 [84]. Principally, reforming of hydrocarbons is not a necessity. In the cold gas system, the removal of water from the synthesis gas is done by condensing. The cooler can be integrated with the steam cycle to produce heat [92]. The removal of CO_2 from the synthesis gas can be done with several technologies.

Table 8
Composition of syngas from different gasification technologies [12].

Syngas properties	Allothermal	Autothermal	Entrained flow	Coal/entrained flow
H_2 content (%)	40	26	39	32
CO content (%)	25	20	38	55
CO_2 content (%)	21	35	20	8
CH_4 content (%)	10	13	0.1	0
C_2H_4 content (%)	2.5	3	0	0
N_2 content (%)	1.5	3	3	3
H_2/CO	1.6	1.3	1.0	0.6
LHV (MJ/m^3)	14	12	10	11

Here indicative syngas compositions produced with different technologies are presented. It can be seen that selected technology affects the gas composition. For use in blast furnace, all gases have suitable gas composition when CO_2 and H_2O are removed.
LHV = lower heating value (MJ/m^3).

CO_2 removal technologies can be classified into four categories [93]: chemical/physical solvent scrubbing, adsorption, cryogenic and membrane technologies.

The Selexol technology is based on physical absorption where a non-reactive fluid is used as the absorbing liquid. The stripping of the solvent is based on pressure change. High pressure and low temperature are utilized in CO_2 removal [94]. Pressure swing adsorption (PSA) is one the most used technologies utilized in industries for gas separation [95].

In Fig. 6**,** a schematic flowsheet for the production of reducing syngas is presented. Pre-treatment indicates grinding and drying of the biomass feedstock. In the gasification step, the biomass is converted into syngas that is cleaned and conditioned further for use. Before the syngas has an adequate reducing ability, CO_2 and water removal operations have to be conducted.

Commercial biomass gasifiers that could be utilized to produce the quantities of syngas needed in ironmaking blast furnace do not exist yet [96]. The largest biomass gasifier has been built in Finland by Metso with a 140 MW fuel capacity. Circulating bed technology is employed, and the plant uses mainly forest chips as feedstock. The plant was run at full power in November 2012 [97]. The advantage of gasification technology could be the co-gasification of biomass and e.g. coal and petroleum coke [98,99].

3.6. Synthetic natural gas

Synthetic natural gas (SNG) can be produced from biomass feedstock by utilizing a gasifier technology as a first step in the chain. After the biomass has been converted to syngas, the gas is cooled and tars are removed. The gas is then cleaned from pollutants. The CO and H_2 in producer gas are then converted to CH_4, CO_2 and H_2O in methanation unit. Further upgrading includes water and CO_2 removal and compression of gas. [100]

One of the forerunners in Bio-SNG process development has been the Energy Research Centre of the Netherlands (ECN) that has developed the MILENA technology to produce SNG from biomass feedstock. With the MILENA technology, 73% SNG production efficiencies can be achieved [101]. The moisture of the biomass fed to the system should be around 10–20%, which means that a drying section must be integrated into the system [102]. Gasification of biomass can be done with different kinds of gasifier technologies such as entrained flow, circulating fluidized bed and allothermal or indirect gasification [100]. The gas composition differs between technologies. The gas cleaning unit follows the gasifier, which removes the solids from the gas. In the methanation process, CO and H_2 from the cleaned gas are converted into methane by the following reactions:

$$CO+3H_2 \leftrightarrow CH_4+H_2O \qquad \Delta Hr=-217\ kJ\ mol^{-1}$$

$$CO_2+4H_2 \leftrightarrow CH4+2H_2O \qquad \Delta Hr=-178\ kJ\ mol^{-1}$$

The reactions are exothermic and the generated heat can be used in the steam production. In the upgrading phase, CO_2 and H_2O are removed from the SNG. The majority of the water is removed when the gas is cooled and rest is removed in the CO_2 removal stage. There are several commercial technologies to perform the CO_2 removal: PSA, physical absorption (Selexol process) or membrane gas separation [100]. The principle of the SNG production system is presented in Fig. 7.

The composition of SNG is comparable to natural gas. The injection of NG to the blast furnace is well-known practice in ironmaking, which is why the injection of SNG is included in this study.

4. Assessment of biomass utilization at different system levels

4.1. Sustainability assessment of bioenergy systems

Ever since sustainability and sustainable development as terms and concepts have been brought up, there have been attempts to develop sustainability assessment (SA) frameworks and indicators to assist in decision-making towards more sustainable practices [103]. There is an expectation that the utilization of renewable energy and especially biomass energy will continue to increase in Europe and world-wide [104,105]. Sustainability of bioenergy production has been addressed in order to ensure the sustainability of energy production on the whole. One of the most used tools to evaluate bioenergy systems is life cycle assessment (LCA) [106,107]. In LCA, the environmental impacts of all the stages of a product's life are evaluated, from raw materials, through production, use to end-of-life. LCA was used to evaluate first-generation biofuels produced from vegetable oils, corn sugar, etc. These studies indicated that food-based biofuels may not be environmentally sustainable and socially

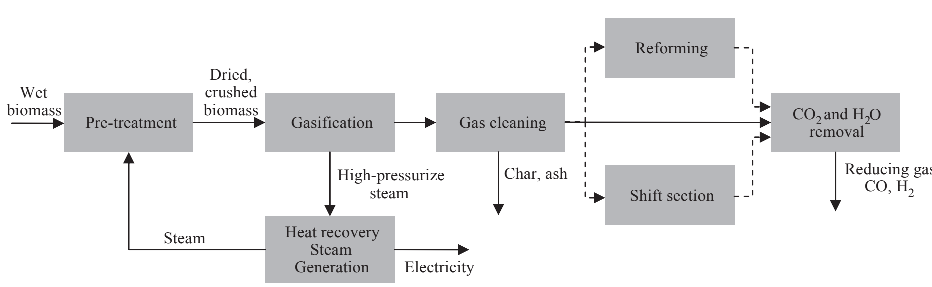


Fig. 6. Simplified process flow chart of syngas production from biomass.

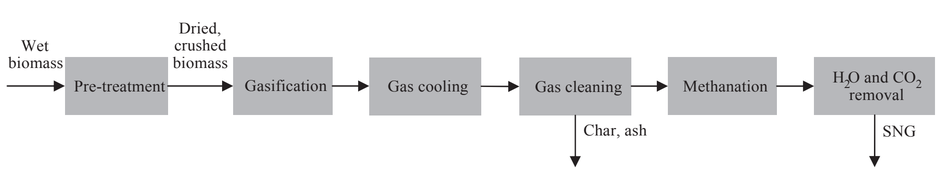


Fig. 7. Simplified process flow chart of SNG production. In reality methanation is complex chain of unit operations where the maximum amount of methane is pursued. SNG=synthetic natural gas.
(modified from [100])

acceptable due to their negative impacts on food security, water and carbon footprints arising from direct and indirect land-use impacts [108,109]. The weakness of LCA as a tool of SA is that it does not consider all the aspects of sustainability.

Several studies have been conducted where different sustainability indicators for renewable energy and bioenergy systems have been proposed e.g. [110–112]. In the forest sector, the assessment evolved from the focus on sustainable wood production to a comprehensive evaluation of environmental, social and economic sustainability of whole value chains [113]. These studies serve as a starting point to evaluate biomass-based reducing agent use in the steel industry. It has been proposed earlier that biomass use in steel industry and its sustainability should be assessed in a bottom-up, layered approach [114]. Firstly, the technological potential of biomass use is evaluated at a unit process level. Secondly, key impacts are assessed on plant and national levels. Finally, supply chain implications are assessed.

4.2. Technological considerations at unit process level (blast furnace)

Similarly to fossil-based reducing agents, biomass-based reducing agents have different ability to replace coke in the blast furnace. A key factor is to evaluate the maximum amount of bio-based reducing agents that could be used in the blast furnace without impairing furnace efficiency and increasing coke rate considerably.

4.2.1. Impacts of charcoal use

Charcoal has very high coke replacement ratio [3,47,115]. This is mainly due to the low ash content and the high share of carbon. Charcoal has low density compared to pulverized coals, which might lead to some difficulties concerning sufficient mass flow rates to blast furnace. However, good combustibility of charcoal and high char burnout ratio are assumed to result in the possibility to use even higher amounts of charcoal than fossil pulverized coal in blast furnace injection [115]. It can be assumed that there are no technological restrictions to use upto 200 kg of charcoal per ton of hot metal in the blast furnace [3,36]. This high charcoal injection rate scenario would result in coke consumption of approximately 260 kg/t hot metal [3], which would be extremely low. In summation, charcoal is an ideal reducing agent for use in the blast furnace.

At the unit process level, the main benefits of using renewable raw material are reduced carbon footprint, better quality of metal and enhanced productivity [3]. Charcoal, produced especially from wood-based feedstock, has a low ash content. In addition, the ash is basic in nature (high Ca) and contains low amounts of inorganic impurities (S and P) [3]. The basicity of ash connotes that there is a decreased requirement of lime or limestone addition to the BF. These two factors result in lower slag amounts in the blast furnace. Babich et al. [3] have calculated that, with charcoal injection, slag amounts could be decreased by 30% compared to pulverized coal injection. Concern has been raised that using charcoal would increase alkali (Na and K) input to the BF [116]. This can be avoided by selecting biomass with low Na and K content.

4.2.2. Impacts of bio-oil use

Ng et al. [117] examined bio-oil injection to the blast furnace with mathematical modeling. Injection of 140 kg/t hot metal bio-oil into the BF resulted in coke consumption of 455 kg/t hot metal, whereas in the base case with pulverized coal injection of 140 kg/t hot metal the coke consumption was 370 kg. From this calculation, coke replacement ratio of 0.25 can be concluded. Bio-oil has high oxygen and water content, resulting in low energy content [118], which weakens its applicability as a reducing agent. The properties of bio-oil resemble that of green biomass, which has been trialed in industrial blast furnace [119]. Wood pellets were injected into blast furnace without any major problems at the rate of 20–30 kg/t hot metal. Coke replacement ratio observed was, however, only 0.35, whereas in the case of charcoal it can be as high as 1.06 [47]. Approximated from heavy oil injection rate [18] and from the assumed coke replacement ratio of 0.25–0.35, coke consumption at 100 kg/t bio-oil injection could be approximately 430–450 kg/t hot metal. It can be concluded that bio-oil is not an efficient reducing agent.

4.2.3. Impacts of syngas and bio-SNG use

Syngas produced from biomass has a varying composition, depending highly on the applied technology and processing conditions. The key factor is the reducing ability. The share of $CO+H_2$ in the gas mix should be well above 90% of the total volume, if gas contains mainly CO, CO_2, H_2 and H_2O. Alternatively, the $(CO+H_2)/(CO_2/H_2O)$ ratio can be used to evaluate the required gas composition. Requirement for this ratio for tuyére injection of syngas is in the range of 11–34 [120]. With proper conditioning, this kind of gas composition can be achieved and coke consumption with syngas injection could be 365 kg/t hot metal [25].

Synthetic natural gas (SNG) produced from biomass feedstock can have similar properties compared to natural gas, thus making the comparison of Bio-SNG and NG rather straightforward. The amounts of NG used in the blast furnaces vary, but the highest rate achieved is 155 kg/t hot metal. This figure is exceptionally high; a more realistic figure would be near 90 kg/t hot metal. Coke replacement ratio of natural gas reported in the literature is 0.9–1.15 [121], which implies a coke consumption of around 365–390 kg/t hot metal.

4.3. Plant level and national sustainability considerations

The key metrics to evaluate the use of bio-reducers at the production plant level are raw material cost, energy efficiency, plant site CO_2 emissions and process/industry integration. At the national level, the assessment includes technology investments, employment and the role of policy instruments. In this assessment, we will focus on steel plant integration, economics of bio-reducers and policy instruments.

4.3.1. Integration of bio-reducer production into the steel plant

There are basically two options for reducing agent production schemes: centralized production and decentralized production [122]. In both cases the integration possibilities with existing infrastructure should be considered. In centralized production, the conversion of biomass into reducing agents would be done in the plant site. In decentralized production, reducing agents are produced in separate production locations and are to be transported to be used in the steel plant. Centralized production of reducing agents can be realized for all the thermochemical conversion processes. Decentralized production would be difficult to establish with gas-based reducing agents. SNG could be transported via NG pipelines, but the coverage of NG network is sparse in many countries. Steel plant process integration crossing industry boundaries has been evaluated in several publications in recent years. Ghanbari et al. [123] have evaluated the economics, CO_2 emissions and energy flows of integration of methanol production plant with integrated steel plant with mathematical optimization. Biomass use as an auxiliary injectant without utilization of pyrolysis by-products has been examined with optimization procedures in [124,125]. In the work of Suopajärvi and Fabritius [6], also possible by-product utilization in heat and power production was estimated. From the viewpoint of biomass upgrading, there are several heat sources available in the steel plant that can be used in biomass drying or as a heat source in thermochemical

conversion [126–128]. The CO_2 profile of the integrated steel plant system can be improved significantly by using charcoal as a reducing agent. Additional decrease in indirect CO_2 emissions can be achieved if external electricity is replaced by electricity produced from pyrolysis plant by-products [6]. Optimal production plant location is dependent e.g. on biomass availability, transportation distances and selected unit size. Especially important are biomass production and supply chain systems, because of low energy density raw material is inefficient to transport [129,130].

4.3.2. The economics of bio-reducers

The economics bio-reducers are greatly affected by the cost structure of the feedstock, applied technologies and other related costs such as energy consumption and labor. The utilization of possible by-products also influences the economics of reducing agent production. However, indicative evaluations of economics can be found in the literature.

The charcoal production cost in Brazil, where short rotation forests are used as raw material, is around 193 €/t charcoal [45]. If it is assumed that the HHV of charcoal is 31 GJ/t the cost of charcoal would be 6.2 €/GJ. The market price of charcoal in the State of Minas Gerais Brazil reached 262 €/t in May 2008 [131]. The cost of charcoal with continuous Lambiotte retort in Australia has been approximated to be in the range of 386 US$/t charcoal [4]. With the assumption of HHV 31 GJ/t charcoal, the cost of charcoal would be 12.45 US$/$GJ_{2009}$. Charcoal production costs from Finnish wood-based feedstock was estimated to be 270–480 €/t charcoal without by-product (gas, liquid) compensation [132].

The cost of bio-oil production without feedstock cost in 40 MW capacity fast pyrolysis plant has been calculated to be 75–150 €/t and 6–12 €/GJ bio-oil depending on the cost data of capital investment [130]. The cost of large-scale production of bio-oil has also been evaluated by Peacocke et al. [133]. The cost range was between 8 and 19.6 €/GJ (130–318 €/t), with feedstock costs ranging from 0 to 100 €/t. The capacity of the plant was 2 t/h. Mullaney et al. [134] compared the production cost of different sized fast pyrolysis plants. Production cost were 320, 261 and 235 US$ per tonne of bio-oil for plant capacities of 100, 200 and 400 metric tonnes of wet biomass per day.

The technologies for production of Bio-SNG are under development and the price for the gas with current technologies is estimated to be 59–97 €/MWh i.e. 16.4–26.9 €/GJ [135]. In the forecast of Energy Research Centre of the Netherlands, the production cost of SNG from biomass could reach the level of 10 €/GJ with large-scale, 1 GW plants in 2030 [136]. In the study by Hacatoglu et al. [137], production cost was calculated for different scales of operation. The highest production cost was 25 US$/$GJ_{2005}$ with small unit size (dry biomass input 500 t/day) and with current technology. The smallest production cost was 16 US $/$GJ_{2005}$ with large unit size (dry biomass 5000 t/day) and with more advanced technology.

The cost of syngas has not been reported in the literature. In the Fischer–Tropsch liquid production process, the pre-treatment, gasification with oxygen and gas-cleaning sections account for 75% of total capital investment [86]. Fundamentally, syngas production is not very different from Bio-SNG production, save from some steps that are excluded. It can be approximated that reducing syngas cost would be about 80% of the cost of Bio-SNG. This would give a cost of 13.1–21.5 €/GJ with the cost range given to Bio-SNG in [135].

4.3.3. Policy and societal impacts

Policy instruments at national and European Union level may also play an important role in the utilization of bioenergy in different industry sectors [138]. The European Union Emission Trading Scheme (EU ETS) might become a powerful instrument to drive industries towards more sustainable operations already in the third phase of EU ETS 2013–2020. In Finland, where renewable energy share in gross final consumption should be 38% in 2020, several instruments related to climate change and environmental policy have been implemented. Examples that steer industrial actors towards the use of bioenergy are energy taxes, investment subsidies, tax refunds, feed-in-tariffs and R&D funding [139]. Energy industry and steel industry might be in uneven competitive situation when considering the use of wood-based biomass as a raw material. The energy industry has received different kinds of subsidies in the past and this trend probably continues also in the future. Subsidies have been paid e.g. for non-industrial private owners, when small-sized wood (thinnings) is used in energy production [140]. This subsidy would probably not be available if small-sized wood was used in reducing agent purposes. Feed-in-tariffs are paid to electricity producers that use forest chips as raw material. The amount of feed-in-tariff is related to peat taxation and cost of emission allowance. Investment subsidies to supply chain machinery and bio-reducer production industry development could also be applied in the case of reducing agent production. This would also result in social benefits, in terms of creating new workplaces.

4.4. Supply chain implications

It was pointed out in Section 4.1 that several issues have to be considered when assessing the sustainability of biomass use. The analysis should be extended to take into account the implications of the whole biomass value chains. In this section, CHG impacts, Energy Return on Investment (EROI) and availability of biomass and competition over raw materials are discussed.

4.4.1. GHG impacts of bio-reducer use

Steel industry is energy intensive and produces vast amounts of greenhouse gases (GHG). Life cycle assessments presented in the literature [141–143] show that 2.16–2.40 t of CO_2 equivalent (CO_2e) are emitted per produced tonne of steel. Two studies can be found where environmental impacts of biomass-based reducing agents in steel industry are evaluated from a life cycle perspective [4,142]. While these studies provide estimates on the net impacts of using charcoal as reducing agent from the viewpoint of iron and steelmaking, there are assessments with LCA view in the literature concerning also other possible biomass-based reducing agents such as charcoal, bio-oil and SNG [69,137,144–148]. Since these biomass-based reducing agents have various energy contents and coke replacement ratios, this section compares the environmental performance (LCA) of thermochemical conversion products as gCO_2/MJ basis without considering coke replacement ratios. In Fig. 8, GHG emissions (gCO_2e/MJ) related to production of charcoal, bio-oil and Bio-SNG are presented [69,137,142,146–149]. There figures can only be held as indicative when comparing the environmental performance of different products, since the underlying assumptions in LCA studies are different. Good example of different assumptions can be seen when comparing GHG emissions from slow pyrolysis of switchgrass A and B by Roberts et al. [69] to other thermochemical conversion routes. In these two cases, indirect impacts on land use have been incorporated, while in the other studies indirect land use has not been taken into account. Calculations are based on lower heating values in [69,137,149] and higher heating values in [147]. In [142,146,148] it was not clearly stated which one was used.

GHG emissions vary significantly between 3.4 and 45 gCO_2e/MJ of product if indirect land use is not taken into account. In switchgrass scenario B, GHG emissions are 119 gCO_2e/MJ biochar.

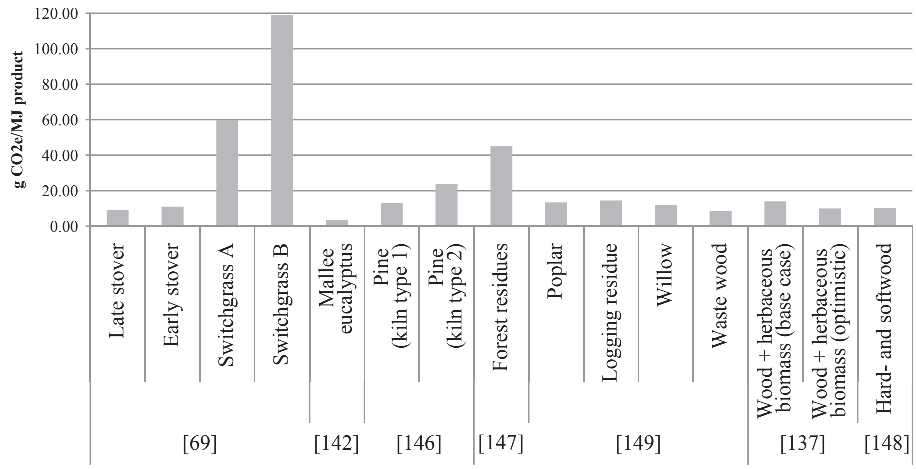


Fig. 8. Life-cycle greenhouse gas emissions (gCO2e/MJ product) of biomass-based reducers reported in the literature and illustrates the GHG emissions from charcoal, bio-oil and Bio-SNG production. They can only be held as indicative when comparing the environmental performance of different products, since the underlying assumptions in LCA studies is different. Feedstock in described studies is also different, which further makes direct comparison difficult. However, comparison shows that GHG emissions are at reasonable level when indirect land use is not incorporated into the calculation.

4.4.2. Energy return on investment

Energy Return on Investment (EROI) can be viewed as an indicator and a numerical quantification introduced to evaluate different energy sources [150]. EROI can be defined as the ratio of energy embodied in the product (primary and by-products) to the energy embodied in materials and processes utilized to produce it [151,152]. The higher the EROI, the more efficiently the energy source is produced. According to the literature review of Raugei et al. [150], the EROI of oil and natural gas have decreased from above 100 to approximately 20. The EROI values for coal are in the range of 40 and 80. Biofuels such as corn ethanol have very low EROI, usually around 1.0 [151]. The EROI of possible bio-reducers was analyzed in some of the assessments [69,137,144,147,153]. Remarkably high EROI, 15–35, depending on the technology, was reported for charcoal [144]. The net energy ratio (a concept similar to EROI) of biochar was 2.8–3.1 in the work of Roberts et al. [69] and 5.3–6.9 in the work of Gaunt and Lehmann [153]. For Bio-SNG, fairly high EROI, 5.1–8.0 was reported [137]. Variations in EROI are due to different production technology and plant capacity. According to Steele et al. [147], production of 1 MJ of bio-oil requires 0.52 MJ energy input taking into account also energy from renewable sources. It is evident that, from the point of view of EROI, there is a true potential in the utilization of bio-based reducing agents.

4.4.3. Feedstock availability and competing uses

Increased demand necessitates the evaluation of the availability of biomass feedstock [111]. The availability of biomass for iron and steelmaking has been evaluated by Piketty et al. in a Brazilian context [154] and by Suopajärvi and Fabritius [132] in Finland. Norgate et al. [142] have estimated the biomass need and required plantation area for mallee eucalyptus plantations when replacing 47% of the fossil reducing agents in integrated BF–BOF-route (990 Mt steel production). The needed biomass amount would be 1500 Mt per year when assuming biomass requirement of 5.2 t/t charcoal. Depending on the plantation yield of (10, 20, 30 t/h/year), the plantation area required is 148, 74 and 50 Mha respectively. Plantation area required for biomass production is significant but, according to Norgate et al. [142], it could be available for biomass production for steel industry. According to our earlier analysis [132], steel industry in Finland would have to compete with other industries, such as energy production in heat and power plants, bio-oil plants and possibly with F-T plants over biomass resources in the future.

4.5. Synthesis of bio-reducer assessments

Table 9 summarizes the advantages and disadvantages of the four potential bio-reducers at different system levels and their assessments related to sustainability.

From a technological point of view, there are various challenges to overcome for bio-reducers being considered a viable alternative. Lower energy content must be offset by other benefits. A thorough sustainability assessment is not available in the literature. Environmental assessments concentrate on GHG balances; however, they do not always take into consideration direct and indirect land-use impacts. On the economic side, the challenge is the high cost of bio-reducer production. Bio-reducers could be economically sustainable should economic incentives be available for biomass use also in the steel industry. Social sustainability assessments of bio-reducers have not been done yet. As a positive asset, biomass-based reducing agent production can have a noteworthy employment impact, as well as knowledge and innovation potential. Feedstock availability studies have been done only in Finland and Brazil. There is uncertainty in availability and conflicts over competing uses could become an issue in the future.

5. The future of bio-reducers; toward a paradigm shift in the steel industry

Steel industry today relies heavily on the use of fossil-based energy in their production processes, with the exception of Brazil, where wood-based charcoal has a strong role as a reducing agent in mini blast furnaces [45]. The prevailing paradigm in the steel industry has been to move toward more environmentally sound operations through enhanced material and energy efficiency, process integration and increased utilization of by-products such as slags, dusts, scales and sludges [126,127,155,156]. Radical technological steps towards large-scale use of new technologies in steel industry have been modest during the era of blast furnace technology, despite the fact that several alternative iron and steelmaking concepts have been developed [157]. Steelmaking based on the use of metal scrap in electric arc furnaces (EAF) is a technology that is more environmentally sound than BF technology. However, increasing demand for steel requires the use of virgin raw materials [157]. Technologies based on the reduction of fine ores without the need of coking and ore agglomeration plants

Table 9
Advantages and disadvantages of the four main bio-reducers.

Bio-reducer Production technology	Charcoal Slow pyrolysis	Bio-oil Fast pyrolysis	Syngas Gasification	Bio-SNG Gasification+methanation
Unit process	*Advantages (Technology)* • Slow pyrolysis technology is uncomplicated.	*Advantages (Technology)* • The production of bio-oil yields char as a by-product that could also be utilized in ironmaking.	*Advantages (Technology)* • Co-gasification of biomass and fossil-based feedstock. • Large coal gasification plants are common. • Could facilitate the use of low quality biomass.	*Advantages (Technology)* • Possibility of co-gasification of biomass and fossil-based feedstock. • High energy yield of product. • Could facilitate the use of low quality biomass.
	Advantages (BF use) • Comparable to fossil PC. • High heating value. • High char burnout ratio • Low ash content. • Low sulfur content. • High replacement rate of fossil fuels in ironmaking. • Increased productivity.	Advantages (BF use) • Fast pyrolysis char may have even better combustion characteristics than slow pyrolysis char	*Advantages (BF use)* • High share of CO and H_2 introduced to the BF has several advantages. • The amount of N_2 decreases in the BF→ Facilitates CO_2 capture if pure oxygen is used with syngas injection. • Almost zero accumulation of unwanted elements (S, alkalis).	*Advantages (BF use)* • Bio-SNG has the same properties as natural gas. • The injection of natural gas is common practice. • Almost zero accumulation of unwanted elements (S, alkalis).
	Disadvantages (Technology) • Typically small-scale. • The largest capacity processes only 25,000 t charcoal per year. • Low carbon and energy yield of product.	*Disadvantages (Technology)* • Large-scale technologies still under development.	*Disadvantages (Technology)* • Needs oxygen/steam as gasification agent. • Large-scale biomass gasification technologies under development.	*Disadvantages (Technology)* • Needs oxygen/steam as gasification agent. • Large-scale technologies under development.
	Disadvantages (BF use) • Achieving sufficient mass flow in injection.	*Disadvantages (BF use)* • High oxygen and moisture content. • Low heating value. • Corrosive • Low coke replacement ratio.	*Disadvantages (BF use)* • The use of reducing gas injection is not proven in large-scale.	*Disadvantages (BF use)* • Large injection amounts might lead to supercooling of the BF hearth.
Plant level and national sustainability considerations	*Advantages* • Production can be integrated with energy production. • Enables decentralized production. • Waste heat from steel plant could be used for biomass drying and pyrolysis.	*Advantages* • Waste heat from steel plant could be used for biomass drying and pyrolysis. • Enables decentralized production.	*Advantages* • Waste heat from steel plant could be used for biomass drying. • Excess high exergy heat available.	*Advantages* • Waste heat from steel plant could be used for biomass drying. • Excess high exergy heat available.
	Disadvantages • Production cost is high. • Charcoal has low energy density.	*Disadvantages* • High production costs.	*Disadvantages* • Decentralized production difficult. • High production costs.	*Disadvantages* • Decentralized production difficult. • Extremely high production costs.
	• The use of biomass in energy production is supported by several incentives at national level, which may not be available if biomass is to be used as raw material in steelmaking. • CO_2 costs or taxes might have strong role in the future, which also has impact on the competitiveness of steel sectors in different regions and countries.			
Supply chain implications	• GHG emissions might be decreased even when the whole life cycle of the bio-reducer is taken into account. • EROI calculations presented in the literature are encouraging. • Challenges of biomass availability and sustainable use in a large-scale. • Conflicts over competing uses might be severe in the future.			

have also been developed [158]. However, the coal or natural gas consumption of these technologies is rather large [16].

Ultra low CO_2 steelmaking (ULCOS) can be seen as a part of the paradigm shift in the steel industry toward entirely different production technologies compared to blast furnace technology [159]. The ULCOS research programme started in 2004, with the aim of reducing CO_2 emissions per tonne of steel by at least 50% compared to the best routes at the time [160] by the use of the

following technologies: (1) blast furnace with top-gas recycling, (2) HIsarna, a new smelting reduction process, (3) advanced direct reduction and (4) electrolysis of iron ore. Additionally, carbon capture and storage (CCS) and sustainable biomass use were defined as supporting technologies [160]. However, the reality in the iron and steelmaking industry is that blast furnace technology will still be dominating hot metal production for a long period of time [157].

The most probable scenario in the future is that bio-reducers will replace only a part of the fossil-based reducing agents [115,132]. For example, in the case of solid injectant, pulverized coal and charcoal could be used as a mixture whereas the amount of each is determined by economics and required chemical properties. Gasification technology could also provide flexible use of renewable and fossil-based feedstock. Co-gasification of biomass and coal is a technically feasible option [98].

The iron and steelmaking sector has not been viewed as a potential large-scale user of biomass in the near future by Taibi et al. [161]. However, according to the economic model of Bellevrat and Menanteau [162], bio-based reducing agent use in blast furnace may be utilized to a great extent in the future, if economic incentives in the form of carbon allowances would take place. In recent research papers [163–165], the cost effectiveness of implementing some of the Best Available Technologies (BAT) and Innovative Technologies (IT) to improve energy efficiency and to reduce CO_2 emissions in the European steel industry have been analyzed. It was concluded that only moderate savings in specific energy consumption and specific CO_2 emissions can be achieved with BAT and IT available in the near future (2030) under different fuel prices and CO_2 emission allowance costs. Biomass was not considered in these studies.

Even though this literature review revealed that the use of biomass-derived reducing agents in blast furnace could be technologically viable, trial periods in large-scale pilot facilities, such as LKAB´s Experimental Blast Furnace (EBF) in Luleå [166] and industrial trials in large modern blast furnaces would be needed. The economics of using charcoal or other bio-based reducing agents in steel industry is one of the most important factors requiring more work. There are several alternatives for process integration, which should be systematically evaluated with thorough techno-economic evaluations. One of the major uncertainties is related to future development of EU ETS, accompanied with national subsidy policies. The ability of different industries to pay for biomass depends on the value of the final product, but also on the subsidies received from the government. More attention should be placed on sustainable growing and harvesting of biomass in the future. Land use changes and total carbon balances need to be thoroughly assessed, to ensure that there will be a net environmental benefit when using biomass.

6. Conclusions

Biomass utilized as raw material in blast furnace ironmaking has a very promising potential to contribute to more environmentally sound steelmaking. Four thermochemical conversion routes for producing reducing agents from biomass feedstock have been systematically reviewed by evaluating the main implications on unit process (BF) behavior, process integration possibilities, economic performance and value chain assessments related to sustainability, including life-cycle GHG emissions and EROI comparisons. The blast furnace process performance with different bio-reducers; charcoal, bio-oil, syngas and SNG was examined based on existing knowledge of comparable fossil-based reducing agents and preliminary evaluations found in the literature.

The study indicates that, from the ironmaking perspective, the most promising renewable reducing agent scenario could be based on the use of charcoal in blast furnace. The replacement ratio of fossil-based reducing agents achieved with charcoal is the highest, thus leading to the largest decrease in fossil-based coke consumption. The amount of injected charcoal into the BF per produced ton of hot metal could be around 200 kg, implying a coke rate of 260 kg/t hot metal. Charcoal production technologies can be kept rather uncomplicated and also the integration with heat and power applications is straightforward. On the other hand, yield of charcoal from slow pyrolysis is low and much of the chemical energy of biomass goes to pyrolysis liquids and permanent gases. Fast pyrolysis bio-oil does not have suitable properties for BF injection, but can be used in heat and power production as such and, with further upgrading, in several other applications. The by-product char (15–25 wt%) from fast pyrolysis could also be utilized in reducing agent applications. Solid and liquid bio-based reducing agents can be produced in a decentralized manner, whereas gaseous bio-reducers require a centralized production scheme.

Thermochemical conversion of biomass to produce bio-reducers seems to have moderate environmental impacts when GHG emissions resulting from indirect land use are not incorporated in the calculations. EROI of bio-reducers found in the literature are well above one (2.8–35), which implies that more energy is gained than what has been used to produce the new unit of energy.

The use of wood-based biomass as a feedstock in bio-reducer production could pave the path for achieving national renewable energy targets set by the EU or other decision-making body. A more in-depth analysis should be performed, in order to evaluate the cost structure of reducing agent production routes, the effect of policy measures and emission trading scheme. Conflicts arising from multiple use of resources ought to be avoided and solutions should be found to ensure a maximum value-in-use for different scenarios. Despite the growing interest in biomass-based reducing agents in the steel industry, more systematic efforts are needed to realize a shift from fossil-based reducing agents to bio-reducers.

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